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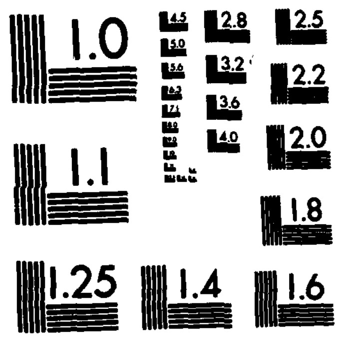

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1. REPORT NUMBER: 5	2. GOVT ACCESSION NO.: AD-A124633
3. RECIPIENT'S CATALOG NUMBER	
4. TITLE (and Subtitle): Reading and Writing of Photochemical Holes Using GaAlAs Diode Lasers	5. TYPE OF REPORT & PERIOD COVERED: Technical Report
	6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s): P. Pokrowsky, W. E. Moerner, F. Chu, G. C. Bjorklund	8. CONTRACT OR GRANT NUMBER(s): N00014-81-C-0165
9. PERFORMING ORGANIZATION NAME AND ADDRESS: International Business Machines, Dept. K46, 5600 Cottle Road, San Jose, California	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: NR 421-001
11. CONTROLLING OFFICE NAME AND ADDRESS: Office of Naval Research, 800 N. Quincy Street, Arlington, VA 22217 Code 240	12. REPORT DATE: February 7, 1983
	13. NUMBER OF PAGES
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	15. SECURITY CLASS. (of this report): Unclassified
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18. SUPPLEMENTARY NOTES

Submitted to Optics Letters

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Optical storage, optical memories, cryogenics, tunable lasers, diode lasers, photochemistry

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A current tuned GaAlAs diode laser is utilized both to burn and to detect narrow photochemical holes in the inhomogeneously broadened 833 nm zero phonon line of the R' color center in LiF. Applications for reading and writing data into frequency domain optical memories based on photochemical hole burning are discussed.

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OFFICE OF NAVAL RESEARCH

Contract N00014-81-C-0165

Task No. NR 421-001

TECHNICAL REPORT NO. 5

Reading and Writing of Photochemical Holes Using GaAlAs Diode Lasers

by

P. Pokrowsky, W. E. Moerner, F. Chu, and G. C. Bjorklund

Prepared for Publication

in

Optics Letters

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February 7, 1983

READING AND WRITING OF PHOTOCHEMICAL HOLES USING GaAlAs DIODE LASERS

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ABSTRACT: A current tuned GaAlAs diode laser is utilized both to burn and to detect narrow photochemical holes in the inhomogeneously broadened 833 nm zero phonon line of the R' color center in LiF. Applications for reading and writing data into frequency domain optical memories based on photochemical hole burning are discussed.

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There has recently been considerable interest in the cryogenic phenomenon of photochemical hole burning[1] both as a tool for high resolution solid state spectroscopy[2,3] and as a means for frequency domain optical storage.[4,5] This phenomenon occurs whenever persistent photochemical changes can be induced by exposure to narrow band optical radiation tuned to a frequency within the inhomogeneously broadened zero phonon line of a photoactive material contained in a crystalline or an amorphous host. The narrow band radiation selectively excites that fraction of the molecules of photoactive material whose local environment is such that their resonant absorption frequency is within a homogeneous linewidth of the excitation frequency. The ensuing photochemical reaction alters the excited molecules so that they no longer contribute to absorption at the laser wavelength, resulting in a persistent narrow hole or dip in the lineshape. Typical ratios of inhomogeneous to homogeneous widths are on the order of 10^3. Thus, 10^3 resolvable holes can be burned into the inhomogeneous line at each spatial location in the sample. If one bit of information is encoded by the presence or absence of a hole at a given location in frequency space and if the exciting laser is focused to a diffraction limited spot 1 μm in diameter, spatial storage densities of 10^3 bits/μm^2 or 10^{11} bits/cm^2 might ultimately be possible.

In this paper, we report the first demonstration of photochemical hole burning using GaAlAs diode lasers. These lasers have many advantageous properties for optical storage applications. They are compact, reliable, have modest power requirements, operate in a single longitudinal and transverse mode with no external cavity, and can be rapidly tuned by varying the injection current.[6] In our experiments, a current-tuned commercially available GaAlAs diode laser was utilized both to burn (write) and detect (read) persistent narrow photochemical holes in the 0.4 nm wide inhomogeneously broadened 833.0 nm zero phonon line of the R' aggregate color center contained in Mg^{++} doped LiF. Exposures on the order of several J/cm^2 were sufficient to write 500 MHz (0.001 nm) wide holes with a depth equal to approximately 5% of the inhomogeneous lineshape.

The R' in LiF:Mg^{++} system was chosen because photochemical hole burning in this system has recently been demonstrated using actively stabilized IR dye lasers.[7] The R' center is not actually a molecule, but rather is a negatively charged intrinsic aggregate color center consisting of 3 trigonally-arranged anion vacancies surrounded by 4 trapped electrons. The center is produced in LiF:Mg^{++} by x-ray irradiation at room temperature.[8] Doping with Mg^{++} ions in the melt has been shown to enhance the achievable R' population.[9] When cooled to liquid He temperatures, irradiated crystals exhibit a prominent zero phonon line centered at 833.0 nm. The photochemical mechanism responsible for the hole burning is conjectured to be ionization caused by electron.tunneling from photoexcited centers to nearby traps.[3,7] Preliminary materials studies[7] suggest that the hole-burning mechanism may also involve a weak photophysical component.

The sample used in our experiments consisted of a 0.6 cm × 0.5 cm × 0.3 cm thick crystal of LiF doped with 0.05 mole % MgF_2 and exposed to Cu x-rays for 100 hours at room temperature. Immediately after x-ray irradiation, the sample was cooled to 2K, and the 833 nm zero phonon line observed in absorption using white light illumination and a spectrometer. The peak absorption at the linecenter was 30% for a 0.3 cm pathlength and the observed inhomogeneous linewidth was 0.4 nm (full width at half-maximum absorption).

The setup for the GaAlAs diode laser hole burning experiments is shown in Figure 1. A single Mitsubishi 3001 TJS GaAlAs diode laser was utilized in all of the experiments. The laser output beam was collimated by a microscope objective and then focused onto the crystal sample which was fixed on a mount contained in a liquid He immersion dewar at 2°K.

Pinholes of various diameters between 50 and 500 μm were mounted directly in front of the crystal to produce a controlled spot size. Intensity and exposure times were adjusted by a shutter and attenuators before the dewar. The power of the transmitted beam was monitored with a fast PIN photodiode (Motorola type MRD510).

Coarse wavelength tuning was provided by temperature tuning. The GaAlAs diode was mounted on a Peltier Cooler which could be adjusted between 0° and 50°C with a stability of 0.05°C. Fine tuning was provided by varying the injection current using specially built electronics. For writing holes, the injection current was stabilized to ± 0.02 mA, resulting in a measured laser emission jitter bandwidth of 200 MHz. For reading holes, the injection current was periodically linearly ramped in a sawtooth manner and at the same time rapidly modulated with a low amplitude, sinusoidal waveform. The ramping caused the laser frequency to repetitively scan over the spectral region containing the hole, while the sinusoidal modulation caused a rapid dithering of the laser frequency for derivative spectroscopy.

The ramping rate was varied from less than 1 Hz to over 20 kHz. Under these conditions, such lasers can be smoothly tuned over impressive ranges. In fact, 60 GHz of tuning without a mode hop has been achieved with slow ramping and 40 GHz of tuning has been achieved at 25 kHz ramping rates.[10] The sinusoidal modulation was performed at a frequency of 2.5 kHz for the slow ramping experiments and at frequencies above 10 MHz for the fast ramping experiments. Derivative spectroscopy of the holes was performed by monitoring the transmitted laser power and detecting the amplitude of the Fourier component of the signal at the sinusoidal modulation frequency. For slow ramping, the 2.5 kHz signals were detected using a lock-in amplifier and displayed on an x-y recorder. For fast ramping, the high frequency signals were homodyne detected using a Mini-Circuits type ZFM-4 double balanced mixer and an oscilloscope for display.[10] In both cases, the displayed signals should show the derivative of the absorptive profile, however, at fast ramping speeds the derivative lineshapes were distorted due to the limited 1 MHz bandwidth of the oscilloscope amplifier (Tektronix 7A22 Differential Amplifier).

The best signal to noise was obtained at slow ramping rates. Since the bandwidth of the detection electronics could be narrowed to less than 100 Hz, the optical detection system was very sensitive. All of the slow ramping experiments were done with the laser power attenuated to 1 μW or less. This prevented deleterious additional hole burning while reading and allowed holes to last for several hours under conditions of continuous repetitive reading. Figure 2 shows typical derivative spectra taken before burning a hole (upper trace), after burning one hole (middle trace), and after burning one additional hole (bottom trace). All three traces were taken at one spot on the sample. For each trace, the laser injection current was slowly ramped between 19.6 and 20.6 mA, resulting in a 9 GHz optical frequency scan. The focused spot size on the sample was 0.002 cm^2. The holes were burned by stabilizing the injection current to a constant value, turning off the sinusoidal modulation, and removing the attenuators to expose the sample to 2 mW of laser power for 15 seconds.

The derivative spectra of the holes is somewhat more complicated than would have been expected for a simple Lorentzian lineshape, since each hole appears to be accompanied by several weaker "sideholes." (Similar sideholes were observed in the IR dye laser experiments of Reference 7.) The predominant central feature is always centered at the burning wavelength. Neglecting the sideholes, an "effective holewidth" can be defined as the frequency displacement between the extrema of the central feature. In these experiments, the effective holewidth was 500 MHz (or 0.001 nm). A comparison of the middle and bottom traces shows that the depth of a previously written hole decreases if a new hole is burned next to it. This "hole interaction" effect is quite reproducible, but does not pose a serious problem for the optical storage application, since the effect becomes much less significant for widely separated holes.

Optical storage applications require data rates which can only be achieved using fast ramping. Figure 3 shows signals which were taken before burning a photochemical hole, after burning one hole and after burning a second hole. (The hole burning conditions were essentially the same as for Figure 2.) However, in this case, the laser frequency was scanned over 17 GHz at a 30 kHz rate, corresponding to a slewing rate of 1 GHz/μsec. The sinusoidal modulation frequency was 179 MHz and the reading laser power was about 0.5 mW. The signal lineshapes are considerably distorted from the true derivative shape due to the limited bandwidth of the detection electronics. This, however, is not a disadvantage for reading data from an optical storage device, since the only relevant information is the presence or absence of the hole. The fastest detection time we could achieve for these relatively shallow, 500 MHz wide holes, was 500 nsec. Under these conditions, the hole could be detected for at least 5 minutes, corresponding to 10^7 reads, before deleterious additional hole burning became significant.

Finally, a series of experiments was conducted to determine the required optical energy density exposure necessary to burn detectable photochemical holes. The focused spot size was carefully controlled by placing a thin foil with pinholes of known diameter directly against the front surface of the crystal. The total laser power incident on the crystal was determined by measuring the power transmitted by the pinhole, crystal, and dewar and then correcting for the known values of the crystal loss and dewar window losses. The energy density exposure was then controlled by varying the exposure time over the range 0.1 to 10 seconds. Figure 4 shows results for an 125 μm diameter pinhole. There is an approximately linear relationship between the derivative signal amplitude and the logarithm of the exposure. It can be seen that very high exposures of several J/cm^2 are necessary. For practical optical storage applications, where detectable holes must be written in 100 nsec or less with a nominal 3 mW of laser power and nominal spot size of 10^{-7} cm^2, this sensitivity would have to be improved by 3 orders of magnitude.

In conclusion, we have demonstrated both writing (burning) and reading (detection) of narrow photochemical holes using practical, current tuned GaAlAs diode lasers. The recording material consisted of R' aggregate color centers contained in Mg^{++} doped LiF. The effective holewidth was about 500 MHz (0.001 nm), implying that 500 resolvable holes could be written into the 250 GHz (0.5 nm) wide zero phonon line at 833.0 nm. We are currently searching for means to enhance the sensitivity of this material to achieve the writing speeds necessary for optical storage applications.

This work was partially supported by the Office of Naval Research.

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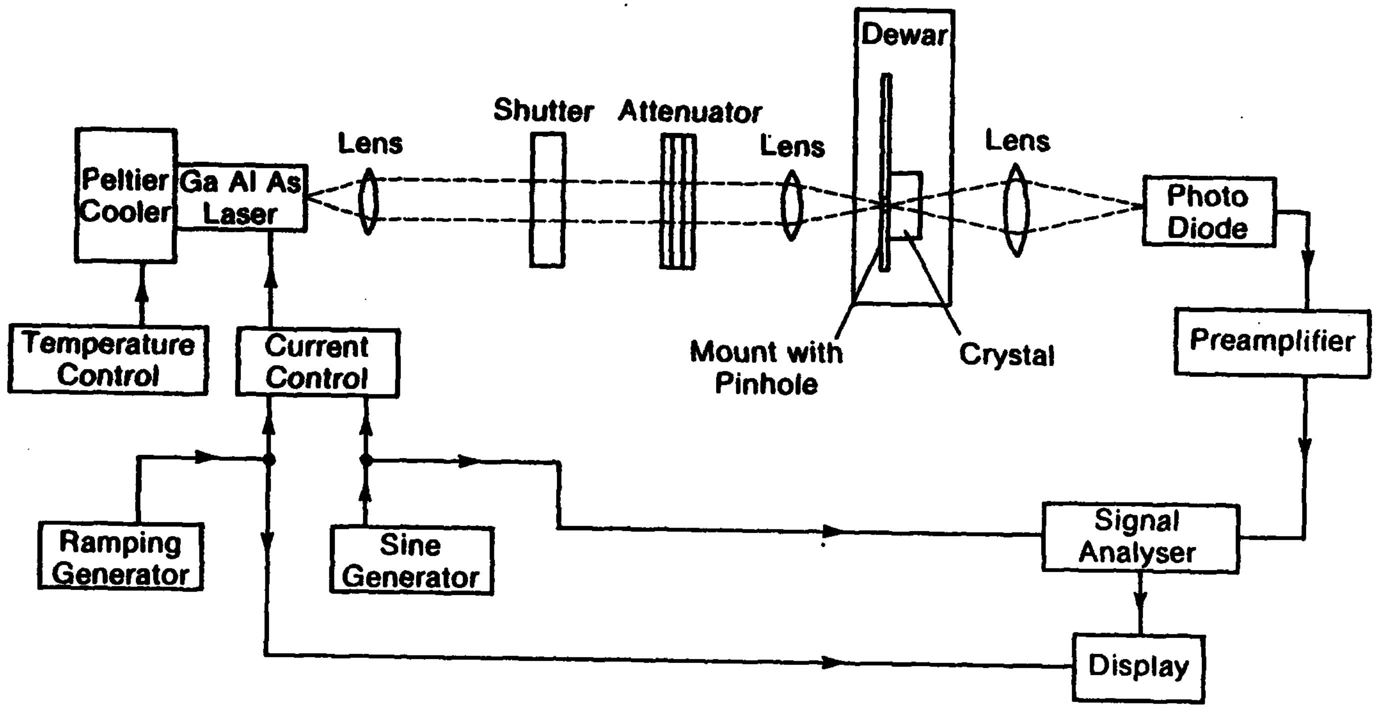

Figure 1. Experimental setup for reading and writing of photochemical holes with a GaAlAs diode laser.

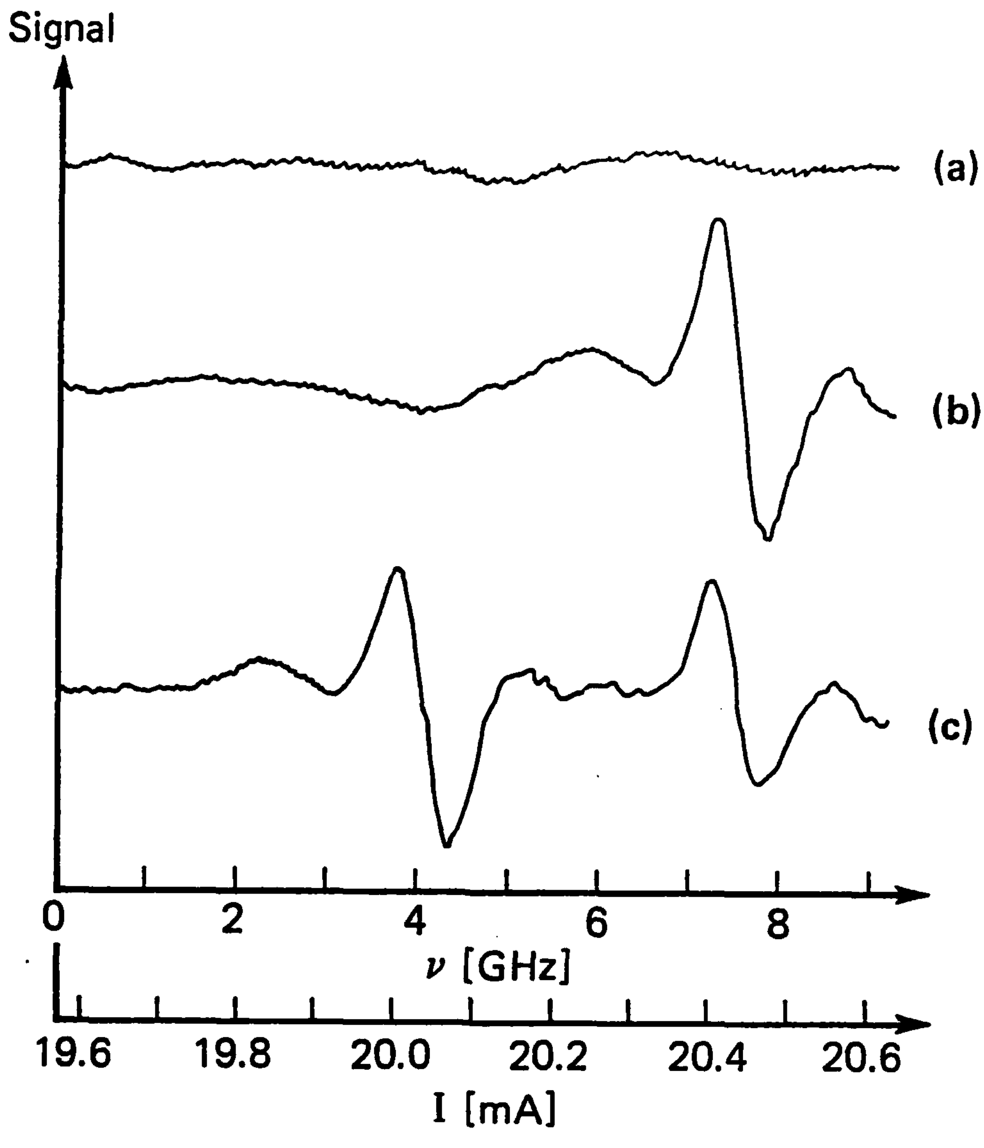

Figure 2. Reading of holes using derivative spectroscopy and slow ramping: (a) before writing a hole, (b) after writing one hole, and (c) after burning a second hole.

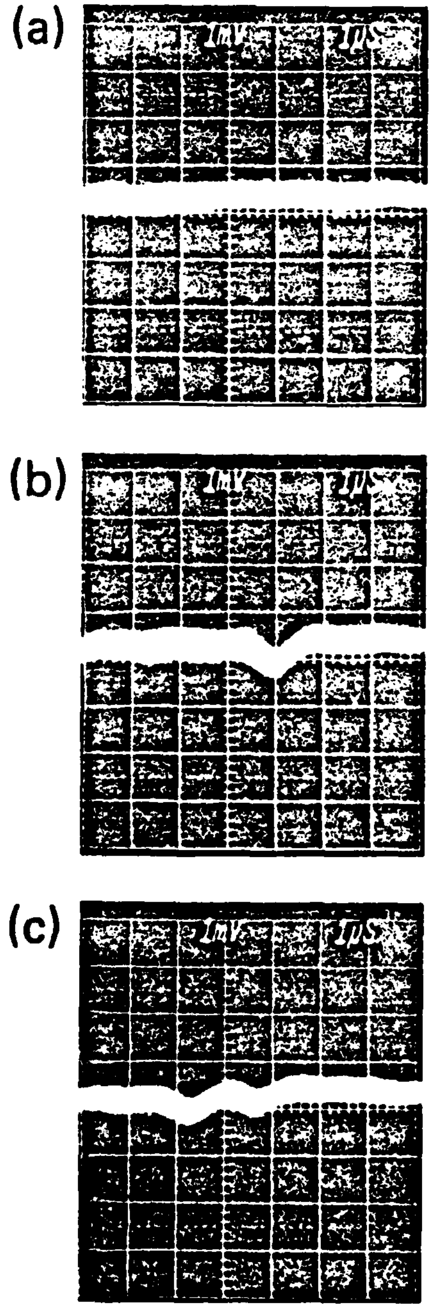

Figure 3. Reading of holes with fast ramping: (a) before writing a hole, (b) after writing one hole, and (c) after writing a second hole.

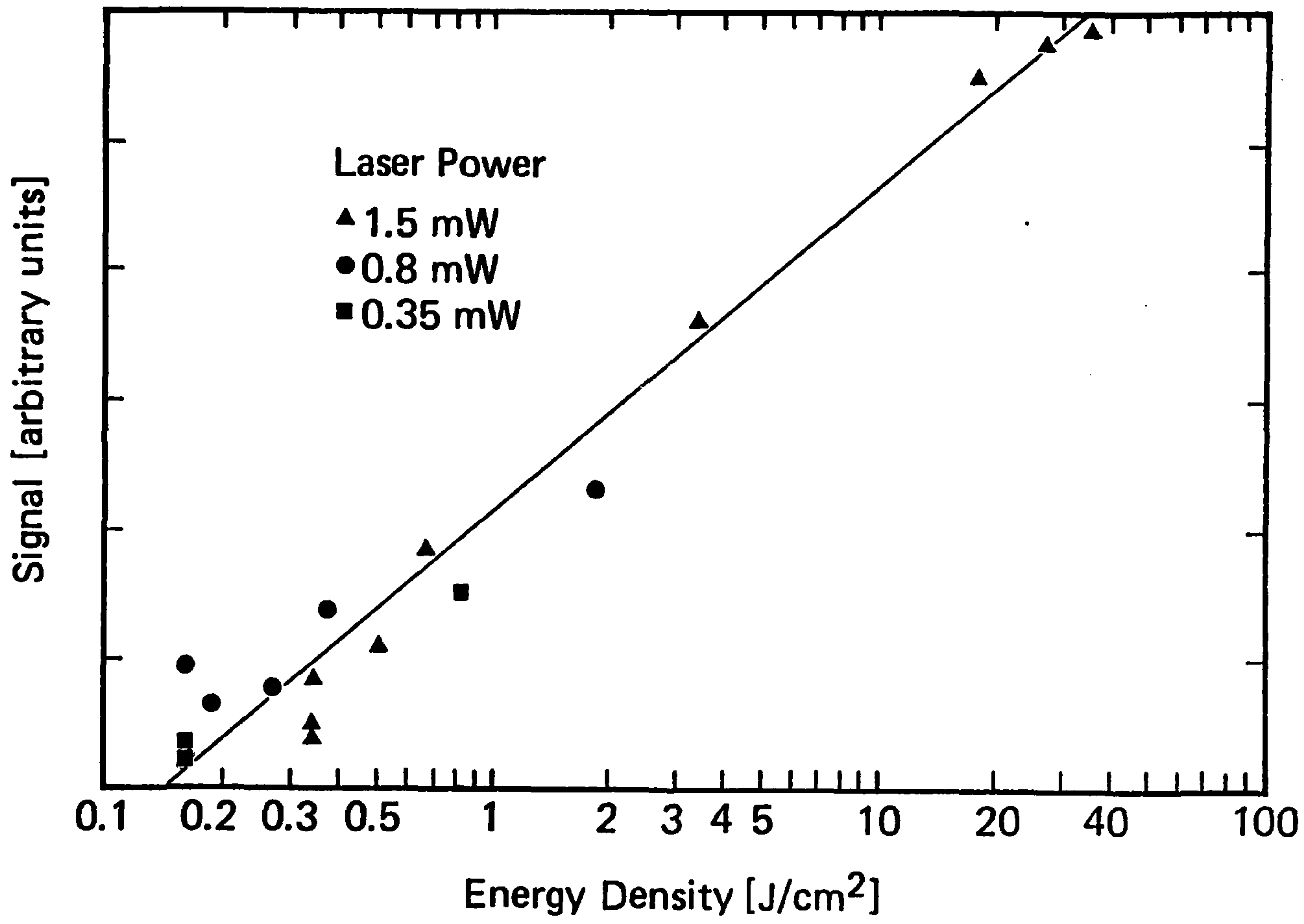

Figure 4. Reading signal strength of holes as a function of the writing optical energy exposure.

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